HOMOGENEOUS OPTICAL SPECTRUM OF CdSe QUANTUM DOTS OBSERVED BY ACCUMULATED PHOTON ECHO

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HOMOGENEOUS OPTICAL SPECTRUM OF CdSe QUANTUM DOTS OBSERVED BY ACCUMULATED PHOTON ECHO

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Temperature-dependent homogeneous optical spectra of CdSe quantum dots was investigated by means of accumulated photon echo. The photon echo decay consists of two exponential decays in the femtosecond and picosecond time domains. The slow decay component manifests the linewidth of the zero-phonon line which is 0.25meV for the 3.6nm-radius dot at 7K, while the fast decay component that of its confined acoustic phonon sideband which is 1.7meV for the dot at 7K. They are much narrower than the linewidth of holes observed by the persistent spectral hole burning. With the increase of temperature, both the linewidths are broadened and Debye-Waller factor decreases. The temperature dependence is explained by the interaction between confined electrons and confined acoustic phonons.

Recently, it has been noted that semiconductor quantum dots (QDs) have very sharp optical spectra due to their atomic-like discrete quantum levels. So far, single quantum dot spectroscopy has observed the spectrometer-resolution-limited luminescence spectra of CdSe and InAs quantum dots [1,2]. An approach to very narrow optical spectra of QDs from the spectral domain faces a difficulty. The homogeneous linewidth of the optical spectra has fruitful information, such as dephasing time and electron-phonon or exciton-phonon interaction at the excited states. Therefore, another approach is required. Photon echo measurement is a time-domain measurement and gives the dephasing time of the excited states. A narrow optical spectral width means a long dephasing time and its measurement is not difficult from the time domain.

The heterodyne-detected accumulated photon echo measurement is known as one of the most sensitive method in the photon echo measurements. The unique requirement for its applicability is the presence of the longer absorption recovery time compared with the laser repetition period [3,4]. Therefore, persistent spectral hole burning (PSHB) phenomena in QDs automatically satisfy the requirement for the applicability of the accumulated photon echo [5]. In fact, we found the heterodyne-detected accumulated photon echo signal in CuCl QDs in glass and NaCl crystals, CuBr QDs in glass and CdSe QDs in glass [6]. It is verified that the Fourier-cosine transform of the time trace observed by the heterodyne-detected accumulated photon echo gives the persistent hole burning spectrum [4].

Samples studied are CdSe QDs embedded in GeO_2:Na_2O glass in the strong confinement regime. They were put into the temperature variable optical cryostat and the accumulated photon echo measurements were done by means of an optical parametric generator pumped by 200kHz regeneratively amplified output of a

femtosecond Ti:sapphire laser. Accumulated photon echo was observed for the lowest energy $1S_{3/2}1S_e$ transition in CdSe QDs under very low excitation density. We observed no power dependence of the echo time trace in the range from $40nJ/cm^2$ to $1.8\mu J/cm^2$.

Figure 1(a) shows temperature-dependent time trace of the accumulated photon echo observed for CdSe QDs (R=3.6nm). The trace was fitted by a linear combination of two exponential decays, $a_f \exp(-t/\tau_f) + a_s \exp(-t/\tau_s)$. The fast decay time, τ_f, was 380fs corresponding to 1.7meV in the spectral domain at 7 K. The slow decay time, τ_s, was 2.7ps corresponding to 0.25meV at 7K. Fourier transform of the time trace shows its homogeneous spectrum of a 0.25meV-sharp line superposed on a 1.7meV-broad band, as is displayed in Figure 1(c) together with the absorption spectrum and the PSHB spectrum shown in Figure 1(b). With the

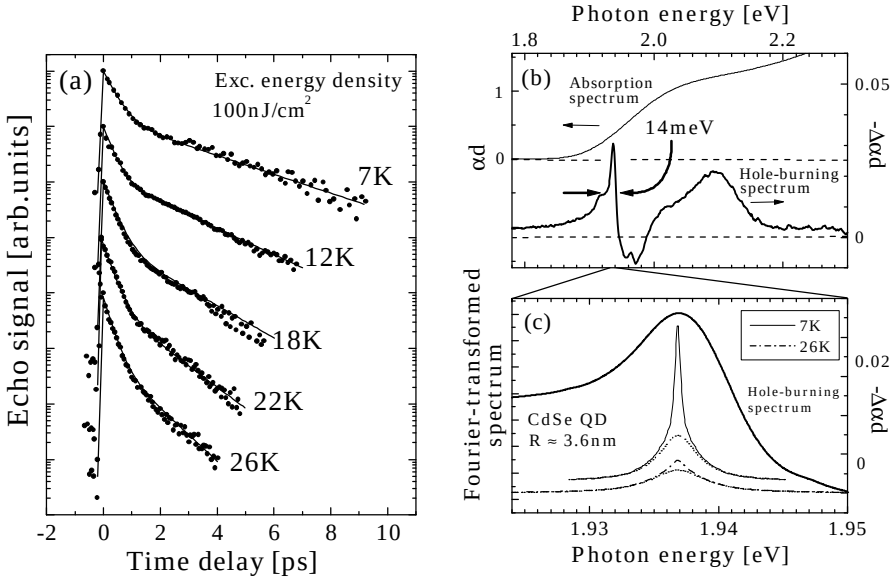

Figure 1. The left figure, (a), shows the accumulated photon echo signal from CdSe QDs (R=3.6nm) under very low excitation density at various temperatures. Solid lines are fitted two-exponential decays. The right upper figure, (b), shows the absorption spectrum (thin solid line) and the PSHB spectrum (thick solid line) of the sample, CdSe (R=3.6nm) QDs in glass. The spectral change was observed at 3min after the burning laser exposure is stopped. The CdSe sample is excited by 9000 shots of dye laser pulses with the photon energy of 1.937eV, and energy density of $0.18mJ/cm^2$. The spectral hole and associated structure are preserved for more than several hours at 2K. The right lower figure, (c), shows Fourier transformed spectrum of the top and bottom time traces of the accumulated photon echo in (a) of CdSe QDs (R=3.6nm) at 7K and 26K. Their broad components are described by dotted lines. The PSHB spectrum in (b) is replotted by a thick solid line.

increase of temperature, the slow decay component, a_s, is suppressed and its decay time becomes faster. The fast decay component, a_f, becomes dominant with the temperature rise. Fourier transform of the echo decay at elevated temperature is also displayed in Figure 1(c). The Fourier-transformed spectrum is much narrower than the persistent hole burning spectrum and the absorption spectrum. The temperature dependence of the slow component contribution, $[a_s/(a_f + a_s)]^{1/2}$ is plotted in Figure 2 and can be well fitted by the expression $\exp[-S_0\coth(h\nu/2kT)]$. This means that the fast component is ascribed to the phonon sideband. On the other hand, the slow component in the echo decay, that is sharp spectrum, is ascribed to the zero-phonon line.

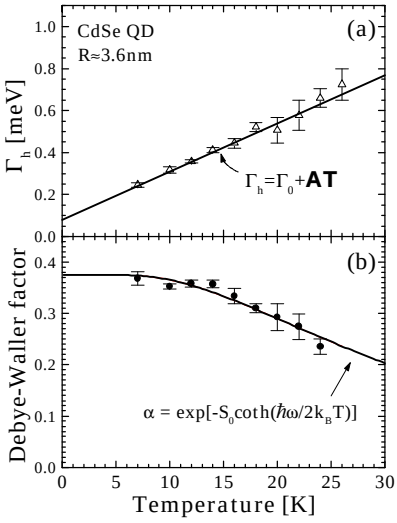

Figure 2. Temperature dependent linewidth of the zero-phonon hole and Debye-Waller factor. Solid lines are fittings by described expressions with parameters, Γ_0=0.08meV, A=0.023meV/K, S_0=0.98 and hν=3.7meV.

Similar spectral features of the homogeneous spectrum consisting of very sharp zero-phonon line superposed on acoustic phonon sideband were observed in CuCl QDs by means of PSHB [7]. It is known that acoustic phonons are confined in QDs and that their energies are inversely proportional to radii of QDs. The experimentally observed confined acoustic spheroidal or torsional mode for CdSe QDs (R=3.6nm) is 1.7meV [8]. It gives the phonon sideband. Further, the fast decay time constant is inversely proportional to radius of CdSe QDs. This observation supports the identification of the fast decay time component to confined acoustic phonon sideband. The width of the zero-phonon line derived from the slow decay increases linearly with the increase of temperature and the linear temperature coefficient of 0.023meV/K almost agrees with the theoretical estimation of 0.017meV/K based on the deformation coupling between confined carriers and confined acoustic phonons fairly well [9]. On the other hand, the width of the broad band, that is, inverse of the fast decay time shows similar temperature dependence.

However, the extrapolated width of the sharp line at 0K is 0.08meV which is much narrower than several meV so far reported in CdSe QDs embedded in glass and is comparable with the result of the single dot spectroscopy of chemically-grown CdSe QDs [1]. The dephasing time constant observed in this accumulated photon echo measurement is much longer than that observed previously in photon

echo measurement [10]. Our measurement was done under weakest excitation condition, so that it is reliable enough. The linewidth observed by PSHB is broader than that of phonon sideband and is much broader than that of the zero-phonon line. This disagreement suggests the spectral diffusion during the PSHB observation is serious. Spectral diffusion may come from variation of the local electric field caused by carrier trapping on the surface of the QDs. Further, built-in local electric field model explains the spectral broadening observed in the electromodutated transmittance spectrum and blue shifted antihole spectrum in the PSHB of the sample.

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